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GROUT TREATMENT FACILITY WASTE FEED ACCEPTANCE CRITERIA

Prepared for WESTINGHOUSE HANFORD COMPANY

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by

D. W. Hendrickson Westinghouse Hanford Company

October 1991

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LIST OF ACRONYMS

DST	Double-Shelled Tanks
SST	Single-Shelled Tanks
GDF	Grout Disposal Facility
GPF	Grout Processing Facility
GTF	Grout Treatment Facility
LLW	Low Level Waste
TRU	Transuranic
DMF	Dry Materials Facility
HWVP	Hanford Waste Vitrification Facility
RCRA	Resource Conservation and Recovery Act
WAC	Washington Administrative Code
TCLP	Toxicity Characteristic Leaching Procedure
LDR	Land Disposal Restrictions
HFW	Hanford Facilities Waste
PSW	Phosphate and Sulfate Waste
DSSF	Double-Shell Slurry Feed
DSS	Oouble-Shell Slurry
NCRW	Neutralized Cladding Removal Waste
NCAW	Neutralized Current Acid Waste
PFP	Plutonium Finishing Plant Aqueous Waste

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WHC-SD-WM-RD-019, Rev. I

GROUT TREATMENT FACILITY WASTE FEED ACCEPTANCE CRITERIA

1.0 EXECUTIVE SUMMARY

This document establishes criteria for the acceptance of grout waste feed to provide assurance that the final grout form produced by the Grout Disposal Facility (GDF) will meet the regulatory, design, product, and process requirements.

Contained in the report is an evaluation of the regulatory requirements associated with the grout disposal option along with a description of the waste currently stored on the site. An evaluation of the heat generation requirements for the waste feed stream is presented. This evaluation includes the heat resulting from the grout curing process as well as heat associated with the radiolytic decay of the radioisotopes present.

Limits for individual elements as well as limits for classes of materials such as organics, sulfates, etc. are presented in Table 1-1. These values are based on regulatory, heat generation, and compositional limits to assure the integrity of the final grout products. Some compositional limits such as heavy metals will require Toxicity Characteristic Leaching Procedure (TCLP) testing to demonstrate regulatory compliance.

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TABLE 1-1: Grout Feed Acceptance Criteria Summary

Feed Component	Acceptable Limit
Organics (ppm) ¹	
TOC	1556
Other Organics	See
	Table 4-2
Cations/Metals (ppm) ²	
Ag	5063
AT	20300
As	0.15
В	136
Ва	46154
Be	TBD-WM-004
Bi ·	TBD-WM-005
Ca	573
Cd	80
Ce	TBD-WM-006
Cr	21000
Cu	7
Fe	1490
Hg	20
K	11500
La	TBD-WM-007
Li	TBD-WM-008
Mg	320
Mn	3010
Мо	68
Na	122000
Nd	TBD-WM-009
Ni	30
Pb	12.5
Pd	TBD-WM-010

TABLE 1-1: Grout Feed Acceptance Criteria Summary

Feed Component	Acceptable Limit
Sb	TBD-WM-011
Se	45
Si	502
Ta	TBD-WM-012
Ti	TBD-WM-013
U .	TBD-WM-014
V	TBD-WM-015
W	TBD-WM-016
Zn	2930
Zr	TBD-WM-017
Anions (ppm) ³	
C1	5360
CN (free)	TBD-WM-018
CN (total)	TBD-WM-019
CO ³	22920
F* ·	562
NO ₃ .	186000
NO ₂	38250
OH-	34850
PO ₄	18430
S0,	. 5100
Radionuclides (Ci/L) ^{4,5}	
H-3	16 μCi/L
C-14	0.647
Co-60	0.1162
Se-79	80.6
Sr-90	0.2662
Nb-94	120.7
Tc-99	0.2617
Ru-106	0.1855

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TABLE 1-1: Grout Feed Acceptance Criteria Summary

Feed Component	Acceptable Limit
Sb-125	0.5399
I-129	0.00107
Cs-134	0.1761
Cs-137	0.3718
Ce-144	0.2237
U-234	
U-235	TBD-WM-014
U-238	
Np-237	
Pu-238	
Pu-239/240	Total TRU concentration <100 nCi/g
Am-241	1200 1101/19
Ст-244	
Other Parameters	
pH (Standard Units)	>10
Total Solids (ppm)	<400,000
Heat Generators	<0.26 CsmBa heat equivalents Ci/L
Density	< 1.4 Kg/L

Notes:

- 1. Total organic constituents should not exceed 3260 mg/L.
- 2. Total sodium (Na) should be greater than 75% of total cations.

 Total aluminum (Al) should be less than 20% of total cations. Waste limitations for As, Ba, Cd, Cr, Pb, Hg, Se, and Ag based on EP toxicity and TCLP tests assuming linearity between waste feed concentration and extract concentrations.
- 3. Total nitrate-nitrite (NO_3-NO_2) should be less than 75% of total anions. Total chloride-fluoride-hydroxide-carbonate $(Cl-F-OH-CO_3)$ should be less than 20% of total anions.
- 4. Performance goal is to limit maximum individual exposure from grout through all pathways to 5 mrem/yr or 0.8 mrem/yr from drinking water.

5. The total mix of radionuclides in the grout feed must be evaluated to assure that the net concentration in CsmBa equivalent curies is 260 per m³. The evaluation method is based on the sum of the fractions rule as described in Hendrickson (1991a).

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2.0 INTRODUCTION

Radioactive liquid and sludge wastes, retrievable by such means as pumping are stored at the Hanford site in twenty-eight double-shelled tanks (DST) and one hundred forty-nine single-shelled tanks (SST). It is the goal of the U. S. Department of Energy (DOE) to provide for permanent disposal of the waste contained in the DSTs. Liquid SST wastes are to be retrieved, pretreated as necessary and placed in the DSTs.

The waste management program at Hanford is described in more detail in the document entitled "Hanford Waste Management Plan (HWMP)." The HWMP (DOE/RL 1988) calls for wastes that have high levels of radioactivity to be processed into borosilicate glass and shipped to the federal geologic repository. The low-level radioactive fraction will be solidified in a cementitious grout at the Hanford Grout Processing Facility (GPF) and disposed in the pre-constructed, lined concrete vaults of the Grout Disposal Facility (GDF).

2.1 Statement of the Problem

The grout resulting from the mixing of the low level radioactive wastes together with the grout forming materials (cement, flyash, etc.) must meet stringent regulatory requirements for such properties as mechanical strength, leachability, thermal stability, and radiation stability. In order to assure that these requirements are met over the design life and/or period of regulatory control of the GDF, the characteristics of the waste feed stream must be well defined.

Wastes contained in the various DSTs and SSTs may contain materials that result in an unacceptable product when mixed with the grout forming materials. In those cases pre-treatment of the waste feed stream may be necessary to alter its makeup. Waste feed materials which may have a potential adverse effect on the resulting grout must be identified and limits established for their composition so that pre-treatment methods can be developed to meet the waste feed acceptance criteria.

2.2 Scope

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This document defines the physical and chemical acceptance criteria for the radioactive liquid and sludge wastes of the DSTs and SSTs, following any pretreatment efforts, for processing, treatment, and disposal in the Grout Treatment Facility (GTF).

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3.0 BACKGROUND

Low Level Waste (LLW) is waste that contains radioactivity and is not classified as high level waste, Transuranic (TRU) waste, mill tailings, or spent nuclear fuel as defined by DOE Order 5820.2A. This definition applies to a broad category of both liquid and solid wastes at the Hanford site. Test specimens of fissionable material irradiated for R&D only, not for the production of power or plutonium, may be classified as LLW, provided the Transuranic (TRU) content of the as-disposed material is less than 100 nCi/g.

Liquid LLW is received from several operating facilities and stored in the DST system. The waste is in the form of a dilute aqueous solution or slurry. The facilities include N Reactor in the 100 Areas; laboratories, T Plant, B Plant, and PUREX Plant in the 200 Areas; and R&D facilities in the 300 and 400 Areas. The 100, 300 and 400 Area wastes are transported by railroad tank cars and unloaded at the 204-AR unloading facility, and can be treated at the facility to conform with DST storage specifications. Except for the nonhazardous phosphate and sulfate waste (PSW) stream, the supernatant associated with these dilute aqueous waste streams, along with other supernatant streams, is evaporated in the 242-A evaporator-crystallizer located in the 200 East Area.

Figure 3-1 represents a schematic of the grout process. A Dry-Materials Facility (DMF) is used to blend the grout-forming solids. The blended solids are combined with the waste in the Grout Processing Facility (GPF) where they are mixed and then pumped as a slurry to the disposal vaults. When monitoring efforts confirm that a stable disposal system exists, a protective barrier system will be placed over the vaults.

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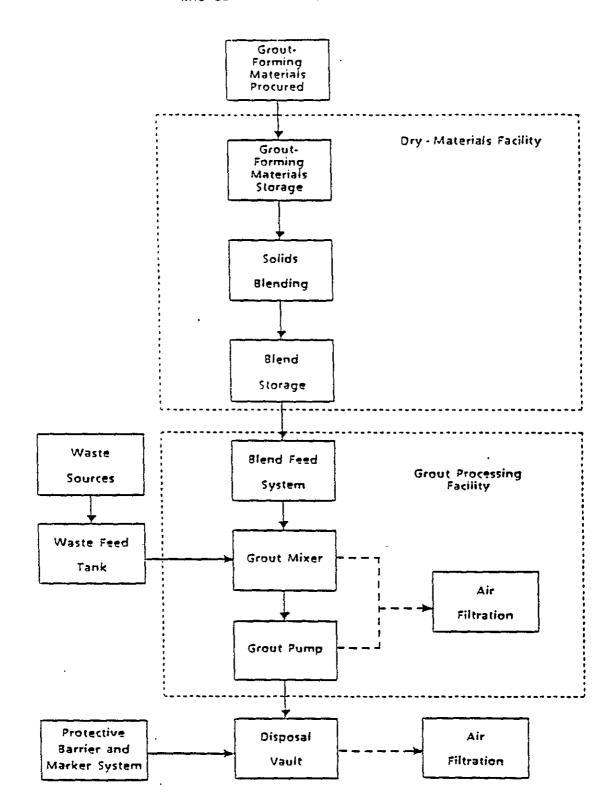
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Several million liters of dilute aqueous LLW are received in the DST system each year. Each stream or batch is chemically adjusted at the source, or possibly at 204-AR in the case of railcar and tanker truck waste, to meet specifications for DST storage. The tank specifications require strict limits for the sodium hydroxide, sodium nitrate, and sodium nitrite content to limit corrosion. It is these chemicals that constitute most of the volume and soluble constituents in these dilute wastes.

These waste streams will be pretreated to separate them into two separate waste streams. The high-level waste (HLW) stream, which contains most of the solids, will be the feed material to the Hanford Waste Vitrification Plant (HWVP); the other stream will be the LLW feed to the GTF. This criteria document defines the physical and chemical requirements for the feed to the GTF.



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Figure 3-1. Schematic of the Grout Treatment Process

4.0 REGULATORY REQUIREMENTS

The grout disposal facility (GDF) consists primarily of near-surface, lined concrete vaults to be used for the disposal of grouted liquid low-level and mixed wastes. These wastes are currently being stored in double shell tanks. The vaults will be managed as surface impoundments and closed as landfills. As such, the facility must ensure compliance with regulations pursuant to the Resource Conservation and Recovery Act (RCRA). These regulations are found in Title 40 of the Code of Federal Regulations (CFR) and Chapter 173-303 of the Washington Administrative Code (WAC) (Ecology 1991). Additional regulatory requirements include those of the Nuclear Regulatory Commission (NRC) and DOE, those pursuant to the Clean Air Act (CAA), and those required within the performance assessment (Whyatt 1991) to assure groundwater quality maintenance.

4.1 Identification of Hazardous/Dangerous Waste

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Mixed waste is any solid waste that contains both a radioactive component and a hazardous (per RCRA) or dangerous (per WAC) component. Washington State also regulates characteristic waste based on WAC toxicity, persistence, and carcinogenicity. Regulations for identifying and listing hazardous/dangerous wastes are found in 40 CFR 261 (EPA 1989a) and WAC 173-303-070 respectively. The radionuclides in the waste are not regulated by RCRA and the WAC.

There are two general categories of hazardous/dangerous waste - characteristic and listed. The double shell tank (DST) waste anticipated for grout feed contains both listed and characteristic waste.

Characteristic wastes are categorized based on ignitability, corrosivity, reactivity and toxicity. Regulations governing designation of characteristic hazardous/dangerous waste are found in Subpart C of 40 CFR 261/WAC 173-303-070. For a discussion of the basis for waste classification and testing see Chapter 3 of the Grout Facility RCRA Part B Permit Application (DOE/RL 1991).

For purposes of preparing grout that will be suitable for disposal, the primary characteristic of concern is toxicity. Table 4-1 gives the maximum concentration of contaminants in a treated waste extract for the characteristic of toxicity based on the Toxicity Characteristic Leaching Procedure (TCLP) for constituents known or anticipated to be in the DST waste.

TABLE 4-1: Maximum Concentration of Contaminants for the Toxicity
Characteristic

EPA HW No. ¹	Contaminant	Regulatory Level (mg/L) 5.0	
D004	Arsenic		
D005	Barium	100.0	
D006	Cadmium	1.0	
D007	Chromium	5.0	
0008	Lead	5.0	
D009	Mercury	0.2	
D010	Selenium	1.0	
D011	Silver	5.0	

^{1.} Hazardous Waste Number (40 CFR §261 and WAC 173-303-090).

The DST waste also contains F003 and F005 listed wastes from non-specific sources in addition to extremely hazardous waste (EHW) concentrations of Washington State Toxic Waste Constituents (WT01). Regulations governing designation of listed wastes are found in Subpart D, 40 CFR §261.

4.2 Disposal Issues

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The land disposal restrictions (LDRs) found in 40 CFR §268 and WAC 173-303-140 provide the basis for determining the standards that the grout feed must meet so that the final product resulting from the grout process will be suitable for land disposal. "Land disposal" for purposes of this document includes placement of the grouted waste in a landfill, surface impoundment or concrete vault.

After the effective date of the LDR, the hazardous/ dangerous waste cannot be disposed in a land disposal facility unless the waste meets the applicable treatment standard, or a variance or exemption applies. Wastes prohibited from land disposal are listed in Subpart C of 40 CFR §268. The F003 and F005 wastes are prohibited from land disposal as are characteristic wastes.

Treatment standards are listed in Subpart D 40 CFR §268. Treatment standards can be expressed as concentrations in waste extract or as specified technologies. Table 4-2 identifies concentrations of the hazardous constituents of F001 - F005 wastes which may not be exceeded for the allowable land disposal of such waste.

TABLE 4-2: Constituent Concentration in Waste Extract ¹				
F001-F005 Spent	Chemical Abstracts Service (CAS) Registry Number	Concentration Limit (mg/L)		
Solvent Constituent		Wastewaters	Nonwastewaters	
Acetone	67-64-1	0.05	0.59	
n-Butyl alcohol	. 71-36-3	5.0	5.0	
Carbon disulfide	75-15-0	1.05	4.81	
Carbon tetrachloride	56-23-5	0.05	0.96	
Chlorobenzene	108-90-7	0.15	0.05	
Cresols (and cresylic acid)		2.82	0.75	
Cyclohexanone	108-94-1	0.125	0.75	
1,2-Dichlorobenzene	95-50-1	0.65	0.125	
Ethyl acetate	141-78-6	0.05	0.75	
Ethyl benzene	100-41-4	0.05	0.053	
Ethyl ether	60-29-7	0.05	0.75	
Isobutanol	78-83 <u>-1</u>	5.0	5.0	
Methanol	67-56-1	0.25	0.75	
Methylene chloride	75-09-2	0.20	0.96	
Methyl ethyl ketone	78-93-3	0.05	0.75	
Methyl isobutyl ketone	108-10-1	0.05	0.33	
Nitrobenzene	98-95-3	0.66	0.125	
Pyridine	110-86-1	1.12	0.33	
Tetrachloroethylene	127-18-4	0.079	0.05	
Toluene	108-88-3	1.12	0.33	
1,1,1-Trichloroethane	71-55-6	1.05	0.41	
1,1,2-Trichloro-1,2,2- trifluoroethane	76-13-1	1.05	0.96	
Trichloroethylene	79-01-6	0.62	0.91	
Trichlorofluoromethane	75-69-4	0.05	0.96	
Xylene		0.05	0.15	
1,1,2-Trichloroethane 2	71-55-6	0.030	7.6 (mg/kg)	
Benzene ²	71-43-2	0.070	3.7 (mg/kg)	

^{1 40} CFR §268.41, Table CCWE, 56 FR 3880, January 31, 1991. 2 Constituent Concentration in Waste, 40 CFR §268.43, Table CCW, 56 FR 3892, January 31, 1991.

Only two of the characteristic wastes identified as F001-F005 spent solvents have technology specific treatment standards; however, no process knowledge of the presence of either 2-nitropropane or 2-ethoxyethanol exists at this time. The treatment technology specified for corrosive waste (D002) is deactivation to remove the characteristic; grout treatment provides such deactivation and thereby requires no waste acceptance criteria based upon disposal. The remaining characteristic wastes have treatment standards expressed as concentration levels; for Table 4-2 constituents, such limits must be met by the waste as generated (Hendrickson 1991c).

The treatment standard for toxic metals is the same as the characteristic level (Table 4-1); testing of waste forms has demonstrated acceptable performance and provides a basis for acceptance criteria. The test results indicate that these levels are achievable through stabilization (EPA 1990).

4.3 DOE and NRC Imposed Specifications for Grout

4.3.1 DOE Order 5820.2A

DOE Order 5820.2A, "Radioactive Waste Management" (DOE 1988), establishes policies, guidelines and minimum requirements for management of radioactive or mixed waste facilities. Chapter 3 contains the requirements for low-level waste facilities that would apply to the management of the grout facility.

Specific requirements include the following limits: 1) external exposure to waste and concentrations of radioactive material which may be released into surface water, groundwater, soil, plants and animals is limited to an effective dose equivalent not to exceed 25 mrem/yr. to any member of the public, 2) atmospheric releases are required to comply with the limits specified in 40 CFR §61 (see Section 4.4) (EPA 1989b), and 3) limits are also imposed on the committed effective dose received by an individual after loss of active institutional controls - 100 yrs.

4.3.2 NRC Limits on Waste Feed

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The radioactive component of the waste feed must be characterized per the requirements of 10 CFR §61 (NRC 1982) to ensure that no waste exceeds the Class C classification limits for radioactive waste. Waste concentrations exceeding the Class C limits are not suitable for near surface disposal and would require a NRC disposal license.

4.4 Clean Air Act Release Limits

The Clean Air Act has requirements and limits on releases of hazardous pollutants to the air. These regulations are generally referred to as NESHAPs and are found in 40 CFR §61 (EPA 1989b). Subpart H of these regulations contains limits for releases of radionuclides to the air from DOE facilities. Emissions shall not exceed amounts that would cause any member of the public to receive an effective dose equivalent of 10 mrem./yr. The regulations also contain monitoring and reporting requirements. Changes in the waste feed would need to be evaluated to ensure compliance with this limit.

4.5 Performance Assessment Limits

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DOE Order 5820.2A (DOE 1988) prescribes that the performance analysis will assure protection of groundwater resources consistent with federal, state and local requirements. To meet this requirement for approval of operations by DOE, performance goals have been developed based on state and federal drinking water protection regulations. These regulations limit exposure to 4 mrem/yr for all radionuclides. The performance goal is a radionuclide dose of 0.8 mrem/yr through the drinking water pathway.

The results of the performance assessment (Whyatt 1991) indicate that the grout disposal system, functioning as designed, will achieve these defined performance goals. Conservative assumptions were made where there was uncertainty in the values to be used for modeling the system. For example, in modeling groundwater transport, the value for dispersion is uncertain so dispersion was not used. Because the impacts of other disposal actions on the groundwater are unknown, the grout disposal performance goals were conservatively formulated using a 20% apportionment of the regulatory limits. Despite the conservative assumptions made in modeling, the performance of the system functioning as designed is still within the performance objectives for all exposure scenarios.

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5.0 WASTE INVENTORY

The wastes managed by the GTF are concentrated salt solutions generated by the operating units in the 100, 200, 300, and 400 areas. Some of the waste is concentrated by evaporation to minimize waste volume. Waste inventories have been developed from existing documentation (Claghorn, 1987; Serne, 1987). The following information provides a brief description of the waste sources, waste stream characterization, waste volumes, and solids contents of the low-level wastes that will be grouted.

5.1 Waste Sources

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5.1.1 Hanford Facilities Waste (HFW) and Phosphate and Sulfate Waste (PSW)

HFW includes the wastes generated on the Hanford site at locations other than the 200 Area operations. The N-Reactor, located in the 100-N Area produced three liquid waste streams. One stream, the N-Reactor decontamination waste, is generated periodically during cleanup operations. Ion-exchange regeneration waste is produced semi-continuously as a result of back-flushing the ion exchange resins used to purify the water in the spent-fuel storage basin. The decontamination waste and ion exchange regeneration waste streams are also known as the phosphate/sulfate waste (PSW).

A third waste stream, the sandfilter backwash waste, was primarily a sludge generated during periodic filter flushing. Other HFW secondary waste streams result from fuel fabrication operations and laboratory activities from the 300 Area, and miscellaneous wastes from the 400 Area.

5.1.2 Double-Shell Slurry Feed (DSSF)

Many streams that enter DSTs consist of dilute liquids low in radioactivity. These streams are so concentrated by Evaporator 242-A that one more pass through the evaporator would increase the sodium aluminate concentration past the sodium phase boundary and the stream would solidify when cooled. At this point the waste is called DSSF.

5.1.3 Double-Shell Slurry (DSS)

When the DSSF is processed through Evaporator 242-A, the DSSF is concentrated past the sodium aluminate phase boundary. The hot slurry is pumped to a DST where it forms solids as it cools. The waste is then called DSS.

5.1.4 Concentrated Phosphate Waste

Concentrated phosphate waste is a blend from different waste sources. Approximately half is phosphate waste derived from N-reactor decontamination operations. The remainder is primarily derived from previous salt well pumping operations. During retrieval, some liquids may be added to facilitate pumping of this waste.

5.1.5 Neutralized Cladding Removal Waste (NCRW)

Cladding removal waste (CRW) results from the dissolution of the N Reactor spent fuel Zircaloy cladding using the Zirflex process in the PUREX reprocessing plant. Neutralization of this waste causes most of the zirconium to precipitate as a hydrated oxide, essentially removing all of the actinides and fission products from the solution. However, sufficient fine plutonium particles are entrained with the precipitated Zirconium that the waste collected in the DSTs is considered to be a transuranic waste. The waste sludge and supernatant as stored in the double-shell tanks is known as NCRW.

5.1.6 Neutralized Current Acid Waste (NCAW)

NCAW is the aqueous high-salt waste from the first-cycle solvent extraction column at the PUREX plant. NCAW contains transuranic (TRU) elements and strontium. The sludge will be separated from the NCAW for disposal. TRU reduced supernatant liquid will be grouted before disposal.

5.1.7 Plutonium Finishing Plant Aqueous Waste (PFP)

The PFP waste originates from the conversion of plutonium nitrate to oxide or metal and includes TRU laboratory wastes. The PFP waste also includes Plutonium Reclamation Facility (PRF) waste consisting of high-salt solvent extraction waste and organic wash waste. Supernatant wastes from the Plutonium Finishing Plant will be disposed in grout following separation of solids.

5.1.8 Complexant Concentrate Waste (CC)

Complexant concentrate waste results from concentration of wastes containing large amounts of organic complexing agents. These organic compounds were introduced to the waste during strontium recovery processing in B Plant.

5.2 Source Term

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The waste inventory in Tanks 241-AN-106, 241-AN-103, and 241-AW-101 have been used to define the compositional range of waste concentrations for double-shell tank (DST) wastes (Hendrickson, 1990). The mean composition of the waste in these tanks is assumed to be representative of the range of waste constituents to be processed by the GTF. This assumption is based on (1) comparisons of sample data with compositions projected from an analysis of process flowsheets, and (2) the expectation that no significant changes in grout feed components will occur over time.

The current DST waste in inventory is primarily material dating from before 1980. Many of the chemical constituents in the current inventory are derived from the salt well pumping program in which residual liquid from retired single-shell tanks were transferred to double-shell tanks. Other waste streams contributing to the inventory are either volumetrically small or otherwise dilute.

Tank 241-AN-106 Wastes

The waste in tank 241-AN-106 (Tank 106-AN) is primarily concentrated phosphate waste from the 100-N Area. The waste was segregated from other tank farm wastes because of the deleterious effects phosphate crystals have on evaporator operations. Other tank waste is salt well liquid and minor amounts of diluted waste.

Tank 241-AN-103 Wastes

The waste in tank 241-AN-103 is primarily salt well liquid. This waste has a higher concentration of aluminate than the other two tanks. The aluminate concentration is indicative of salt well liquids.

Tank 241-AW-101 Wastes

The waste in tank 241-AW-101 is primarily dilute wastes discharged from the PUREX Plant and concentrated in the evaporator. This waste is characterized by high concentrations of potassium in comparison with the other two tanks. High concentrations of potassium are indicative of PUREX wastes in the same manner that aluminate is indicative of salt well liquids and phosphate is indicative of wastes from the 100-N Area. The remainder of the tank waste is salt well liquid and minor amounts of dilute waste.

5.3 Physical/Chemical Characteristics

Appendix A, Tables A-1 through A-5, contain mean, 95% confidence, and bounding source term concentrations for organic, cationic, and anionic species, radionuclides, and other physical parameters. Definitions of these terms are contained within Appendix A. The source term characteristics were based upon samples from Tanks 241-AN-103, 241-AN-106, and 214-AW-101. As discussed in Hendrickson (1990) and Claghorn (1987), these analyses are representative of DSS and DSSF wastes and are expected to bound, following pretreatment, other waste types.

5.4 Volume

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Under current design specifications, each grout vault will contain approximately 3.785 million liters (1 million gallons) of tank waste. Grouted waste occupies approximately 40% more volume than the waste itself. Current facility design and waste volume projections encompass the filling of 43 disposal vaults (DOE/RL 1991). Waste volume data used in preparation of this document are described in Hanlon (1991).

5.5 Trends for Future Waste Feed Component Variations

Future waste streams will include dilute, non-complexed waste from various facilities and B-Plant Aging Waste supernatant from retrieved Aging Waste. A smaller volume of concentrated complexed wastes, NCAW, and NCRW will also be produced.

5.5.1 Dilute/Non-complexed and Aging Waste Supernate Wastes

The character of the dilute/non-complexed and Aging Waste supernatant DST wastes is based on known tank-waste compositions, waste volumes, and anticipated blending operations, as reported in the Grout Facility Part B Application (DOE/RL 1991) and by Claghorn (1987). Due to the comparable solubilities of ¹³⁷Cs and sodium, the data reported by Claghorn (1987) has been normalized to a 5 M sodium concentration to account for the radiolytic heat loading of the waste. Further operational experience indicates that significant precipitation of inorganic waste components may occur at concentrations above 5M sodium. Therefore, it is assumed that wastes will be blended to this concentration to ensure a relatively homogeneous feed.

The composition of these future wastes differs from the current DST waste composition with respect to nitrite-nitrate, aluminate, and chloride. As the waste ages, the ratio of nitrite to nitrate will increase due to radiolytic effects; the current ratio of nitrite:nitrate is approximately 1:1. Total nitrate concentration is expected to be less than 3M (186,000 mg/L). Aluminate concentrations are expected to drop from current levels (0.4M to 0.7M; 25,000 - 43,000 mg/L) to less than 0.3M (18,600 mg/L) due to the cessation of dissolution operations to declad aluminum-clad fuel rods. The highest chloride concentrations are anticipated to be 0.03M (1,000 mg/L) and may be as high as 0.3M (10,000 mg/L). The source waste feeds that are expected to exhibit the highest concentrations for these and other constituents of interest are listed below:

•	NO ₃ -	-	B-Plant	Vessel	Clean-Out	Pretreated	Complexed	Wastes
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- NO₂ B-Plant Cell Drainage Vitrification Plant 222-S Laboratory
- AlO₂ Retrieved PFP Solids Salt Well Liquids
- SO₄⁻² 100-N Sulfate Streams 300,400 Area Waste Fuel Fab
- F Purex Decladding Waste (Post 1987)
- PO_z T-Plant Decontamination Waste
- · CO₃ Purex Miscellaneous Wastes
- Cl B-Plant dilute, non-complexed waste from processing of concentrated complexed waste.
- K⁺ Purex Decladding Wastes
- MnO_a T-Plant Waste

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5.5.2 NCAW and NCRW Waste

The GTF may receive six product streams from the processing of NCAW (Wong, 1989). NCAW sludge containing TRU elements and strontium are expected to be separated before disposal at the GTF. The remaining supernatant may be grouted for disposal. NCAW waste feed to the GTF is expected to contain relatively high concentrations of aluminate and cesium.

The decladding of fuel rods produces a two-phase waste consisting of liquid and sludge. The liquid phase can be separated and retrieved leaving behind a sludge referred to as NCRW. The sludge is expected to contain relatively high concentrations of fluoride, zirconium, and potassium. The NCRW sludge may require modification before retrieval.

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6.0 ANALYSIS OF TEST RESULTS

Grout quality is demonstrated by preparing a sample from actual or simulated feeds. These samples are mixed with an appropriate blend of dry materials and tested for physical characteristics including processability, compressive strength, leach resistance, and TCLP results. The need to demonstrate grout quality is based on the fact that final grout characteristics will vary with changes in feed, process, and formulation compositions.

The success of the product demonstration at the feed tank is dependent upon the success of previous formulation development. The current formulation strategy is to define a waste stream and develop experiments to determine how different mixtures of the dry components affect grout characteristics. To date, ORNL has developed grout formulations for two Hanford feed types: PSW wastes and NCRW supernatant waste. Battelle Pacific Northwest Laboratory (PNL) has been investigating leaching characteristics of different grout mixtures to evaluate the performance of the grout product in retaining hazardous components. The PNL tests have been conducted using PSW, Tank 106-AN, and DSSF waste feed types. The chemical analysis of these waste feeds are summarized in Table 6-1.

6.1 Characterization of PSW Grout Formulation

6.1.1 Laboratory Study

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Leaching and adsorption characteristics of PSW grout was investigated in 1987 (Serne, 1987). Experimental data from three leach tests (ANS 16.1 intermittent solution exchange test, static leach test, and once-through flow column test), two adsorption tests (batch and once-through flow column), and a combined grout-leaching, sediment-adsorption column test were used to (1) characterize the ability of PSW grout to resist leaching of waste constituents to groundwater, and (2) identify mechanisms that control leach rates and adsorption potential.

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TABLE 6-1: Compositions of DSSF, Tank 241-AN-106, and PSW Waste Feeds.					
Constituent .	Simulated DSSF, (mg/L) ¹	TK-106-AN, PNL (mg/L) ²	TK-106-AN, WHC (mg/L) ²	PSW (mg/L) ³	
Ag	162	-	·	_	
Aì	20300	10800	12465	8.1	
As	.03	-	-	< 0.08	
Ва	600	•	-	< .002	
Ćl	5360	2438	3474	220	
Ca	573	70	85	22	
Cd	8	-	-	< 0.004	
Cu ⁺²	7	-	1.5	.5	
SO ₄	5100	2650	2592	2000	
Fe ⁺³	1490	-	-	170	
Mg	320	-	<u>.</u>	-	
Ρ	2020	4400	6260	-	
PO ₄	5653	15225	18430	11600	
Hg	3	-	-	<u></u>	
K	11500	31	32	<0.3	

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TABLE 6-1: Compositions of DSSF, Tank 241-AN-106, and PSW Waste Feeds. Simulated DSSF, $(mg/L)^1$ TK-106-AN, PNL $(mg/L)^2$ TK-106-AN, WHC $(mg/L)^2$ Constituent PSW (mg/L)3 B 136 29 18 CO_3 8970 22920 Cr 1260 662 832 3.5 NO₃ 186000 88500 90024 400 Si 502 55 28 3.9 Zn 2930 NO₂ 22977 38250 36754 Ni 30 27 5 1.5 Pb 2.5 < 0.06 Se 4.5 Si 55 28 8.9 Zn 1616 17 Other **Parameters** pH 12.4

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	TABLE 6-1: Compositions of DSSF, Tank 241-AN-106, and PSW Waste Feeds.							
Const	ituent	Simulated DSSF, (mg/L) ¹	TK-106-AN, PNL (mg/L) ²	TK-106-AN, WHC (mg/L) ²	PSW (mg/L) ³			
Cemer	t Type	47% Fly Ash, 47% Blast Furnace Slag, 6% Type I-II Portland Cement	47% Fly Ash, 47% Blast Furnace Slag, 6% Type I-II Portland Cement	47% Fly Ash, 47% Blast Furnace Slag, 6% Type I-II Portland Cement	41% Type 1-II Portland Cement, 40% Class F Flyash, 11% Attapulgite-150 Drilling Clay, 8% Indian Red Pottery Clay			

Notes:

1. Whyatt (1989), Serne (1989b). Lokken (1988), Claghorn (1987).

1.1 Kg/L

2. Serne (1989b).

Dry Addition

3. Fow (1987); Lokken (1989).

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The grout waste form used in this investigation was simulated to resemble HFW solutions that might result from a decontamination operation (phosphate waste) and a fuels storage basin water cleanup process (sulfate wastes) at the Hanford N-Reactor. The assumed blend of phosphate waste:sulfate waste was 3:2. The phosphate waste was actual N-Reactor waste and contained measurable activities of 55 Mn and 60 Co. The sulfate solution used was a chemically simulated liquid waste spiked with 85 Sr and 137 Cs. Chemical analyses of the grout waste feed were not performed and the presence of secondary constituents were not quantified. Table 6-2 lists the composition of the PSW Grout formation.

TABLE 6-2: Composition of PSW Grout Used in 1987 Leaching/Adsorption Tests

Solids	Portland Type I and II Cement	41 wt%
	Class F Fly Ash	40 wt%
	Attapulgite Clay	11 wt%
	Indian Red Pottery Clay	8 wt%
Liquids	Sulfate Waste Components	40 wt%
	0.03 M Na ₂ SO ₄	
	0.01 M NaOH	•
	0.02 M NaNO ₂	
	Phosphate Waste Components	60 wt%
•	0.151 M Na ₃ PO ₄	•
	0.013 M NaNO ₂	
	0.01 M NaOH	

Although informative, this investigation is not directly applicable to the development of waste specifications for the following reasons:

- The test results did not include grout acceptance criteria parameters,
- complete chemical analyses of specific waste feeds are not readily available, and
- chemical characterization of the unsolidified grout are not available.

6.1.2 Pilot-Scale Studies

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A major pilot-scale test produced 83,270 liters of simulated grout was conducted in July 1986 to assess the effectiveness of the grouting operations and the resulting grout properties (Fow 1987, and Lokken, 1988). During the test, 60,560 liters of simulated PSW waste were solidified with a four component blend of dry solids. The solids included portland cement (41%), Class F fly ash (40%), illitic clay (8%), and attapulgite clay (11%). Dry solids were mixed at two ratios: 3.2 and 3.3 kilograms dry mix per gallon of waste. Equal volumes of phosphate waste and sulfate waste were mixed to

produce the waste feed; a small volume of sandfilter backwash sludge was also included in the sulfate waste. The solids present in the sulfate waste were present at a ratio of approximately 50 kg to 1 million liters.

Investigation parameters included rheology, Extraction Procedure Toxicity (EPTOX) of simulated PSW waste and bleed liquid, Toxic Characteristic Leaching Procedure (TCLP) of 22 grout monolith samples, compressive strength of cured grout, drainable liquid fraction, and bulk density. Of these, rheology, EPTOX, TCLP, and compressive strength are directly applicable to grout acceptability for operational needs, RCRA requirements, and NRC Guidelines (NRC/NISTIR 1989). Only inorganic constituents in extracts were analyzed in the EPTOX and TCLP tests. The major findings of the pilot test are summarized below and in Table 6-3.

- The flow characteristics of the grout mixture were determined to be acceptable. Desired turbulent flow through the inlet pipe was observed.
- TCLP leachate analyses were within regulatory limits (Table 6-3).
- The compressive strength of the grout ranged from 258-440 psi.
- Drainable liquid ranged from 3.59-16.4 % (by volume).

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The density of the unsolidified grout was 1.3-1.4 Kg/L.

TABLE 6-3: Results of July, 1986 TCLP Tests of PSW Grout

Analyte		TCLP (mg/L)	REG LIMIT (mg/L)
As	<	0.5	5
Ba		0.47	100
Cd	<	0.008	1
Cr		0.04	5
РЬ	<	0.12	5
Нд	<	0.002	0.2
Se	<	0.05	1
Ag	<	0.5	5

6.2 Characterization of Tank 241-AN-106 Grout Formulation

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(C) (C) PNL has also conducted laboratory tests and collected empirical leach rate data for various chemical species (Serne 1987). The species investigated included radionuclides (60 Co, 90 Sr, 99 Tc, 129 I, 137 Cs, 241 Am), stable major ions (NO_3 , NO_2 , F, CI, and Na^+), and trace metals (Cr, Mo, Ni). The grout used in the test was produced by mixing 1080 grams of dry blend with 1 liter of waste from Tank 106-AN (9 pounds per gallon). The dry blend was composed of ground blast furnace slag (47.5 wt%), Class C fly ash from Centralia, Washington (47.5 wt%), and Type I-II Portland Cement (5 wt%). Two types of tests were used to generate leaching data:(1) an intermittent replacement leach test (ANS 16.1 leach test), and (2) a static leach test. In addition, an EPTOX was also performed on a grout sample.

Results (Serne, 1989a) indicate that the leaching characteristics observed exceeded (achieved and surpassed) the waste form criteria. Of the species investigated, ^{99}Tc , ^{129}I , Cl, NO $_2$, NO $_3$, and Na are predicted to have the highest leach rates based on observed diffusion coefficients. Mo is also expected to be a probable contaminant of concern. These results compare favorably with similar tests performed by ORNL on Tank 106-AN grout prepared at a mixture of 8 pounds dry blend to gallon of waste (Tallent, 1988). The predicted leach indices for the five species tested all exceed the acceptance criterion of 6.0 (Table 6-4). The EPTOX test indicated that Tank 106-AN extractant is below regulatory limits (Table 6-5).

TABLE 6-4: Results of ANS 16.1 Leach Tests of Tank 106-AN Grout

Analyte	PNL Data (leach index)	ORNL Data (leach index)
99Tc	7.4 ± 1.2	9.1 ± 0
129I	7.6 ± 0.4	7.8 ± 0.1
NO ₃	8.2 ± 0.5	8.0 ± 0.1
NO ₂	8.1 ± 0.5	8.0 ± 0.1
C1	7.0 ± 0.6	7.7 ± 0.2

TABLE 6-5: _Results of EPTOX Test of Tank 106-AN Grout

Analyte	EPTOX (mg/L)	REG LIMIT (mg/L)
As	<0.25	5
Ba	0.48	100
Cd	<0.01	1
Cr	0.07	5
Pb	<0.10	5
Hg	0.0001	0.2
Se	<0.25	1
Ag	<0.01	5

6.3 Characterization of DSSF Grout Formulation

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Grout leaching tests are currently underway in support of the WHC Grout Disposal Program (Serne, 1989b, and Lokken, 1989) to answer key performance questions concerning extrapolation of laboratory testing to full-scale disposal operations. The tests use simulated DSSF mixed with a three-component dry blend of Type I-II Portland Cement (6%), fly ash (47%), and blast furnace slag (47%). The dry materials are blended with the waste at the ratio 1080 grams dry solids per liter of waste (9 lb per gallon).

Preliminary results indicate that DSSF grout also exceeds the waste form criterion for leachability. These tests also focused on the species 99 Tc, Cr, Mo, Na, NO₂, and NO₃. These tests were conducted using Hanford groundwater and deionized water as leachate solutions. The predicted leach indices for these five species tested all exceed the acceptance criterion of 6.0 (Table 6-6).

TABLE 6-6: Results of ANS 16.1 Leach Test of DSSF Grout

Analyte	Groundwater (leach index)	Deionized Water (leach index)
⁹⁹ Tc Cr Mo Na NO ₂ NO ₃	8.77 ± 0.26 11.07 ± 0.3 8.18 ± 0.25 7.75 ± 0.25 7.81 ± 0.28	8.21 ± 0.09 10.39 ± 0.31 7.91 ± 0.24 7.51 ± 0.26 7.57 ± 0.35
NO ₃	7.61 ± 0.28	7.44 ± 0.35

EPTOX tests were also conducted on 9 grout core samples from a DSSF grout pilot-test (Lokken et al. 1989). All EP toxic metal concentrations in the EPTOX extract are below regulatory limits (Table 6-7).

TABLE 6-7: Simulant DSSF EPTOX Results

Analyte	EPTOX (mg/L)	REG LIMIT (mg/L)
As	<1.0	5
Ba	0.5-1.3	100
Cd	<0.1	I
Cr	0.1-0.3	5
РЬ	<1.0	5
Hg	<0.03	0.2
Se	<0.1	1
Ag	0.06-0.16	5

Tests were conducted on solidified grout made from the PSW, 106-AN, and simulated DSSF wastes. The results of the tests are in Table 6-8. As indicated in Table 6-8, the only tests conducted were leachability, toxicity, and compressive strength (PSW only). The tests conducted were successful and exceeded the suggested criteria (NRC/NISTIR 1989) for the grout made from three wastes as shown in the table.

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TABLE 6-8: Summary of Test Analysis Data

Tests	Methods	Criteria	PSW Test ¹	106-AN Tests ²	Simulated DSSF ³
Compressive Strength (S _c)	ASTM C39 or D1074	60 psi	258-440 ps i		
Radiation Stability		S > 60 psi after 10 ⁸ R			·
Biodegradability	ASTM G21 & G22	No Growth & S _c > 60 psi			•
Leachability	ANS 16.1	Leach Index > 6	Passes	Passes	Passes
Immersion		S _c > 60 psi after 90 days			
Thermal Cycling	ASTM B553	S _c > 60 psi after 30 cycles		•	
Free Liquid	ANS 55.1	0.5%	•		
Full-Scale Tests		Homogeneous and correlates to lab size test results		•	-
EP Toxicity			Passes	Passes	Passes
TCLP			Passes		

Notes:

- 1. Fow (1987); Lokken (1988)
- Serne (1989b).
 Serne (1989a); Lokken (1989).

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7.0 GROUT WASTE FEED HEAT GENERATION ANALYSIS

The primary purpose of this section is to define the heat generation criteria for the grout waste feed to assure that the resulting grout performance requirements are met. Toward this end, many of the "Methods of Determination," which describe how a criterion will be met, are based on pilot-scale experiments or laboratory tests on samples of simulated grouted waste and computer code analysis. The eventual application of these criteria to the full scale grout process will require some definition of process control parameters to assure that the end product will still conform to all the waste form criteria.

7.1 Cure Temperatures

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One of the most critical parameters that affects the acceptability of the grouted waste is the maximum cure temperature. It has been shown experimentally (Fow, 1987) that the grout will have acceptable physical properties when the peak cure temperature is kept below 100°C. Other ongoing work has indicated that long curing periods at temperatures as low as 75°C have resulted in grouts not meeting all criteria. As a result of this work, and as a conservative measure below 100°C, a 10°C margin is used, reducing the peak temperature criteria to 90°C. Two sources of heat are considered in demonstrating compliance to this 90°C peak temperature criteria: heat of hydration and radiolytic decay heat. A thermal analysis (Allen, 1990) of peak temperature profiles has been completed based on a small scale experiment. The radiolytic heat was assumed to be constant (0.12 Btu/hour ft3). This value for radiolytic heat generation agrees well with that derived by Hendrickson, 1990 for scoping analysis. The computer code used is TAPA (Guzek, 1990) which has complied with Westinghouse QA level 2 requirements. The results of the computer analysis are reported in Allen (1990).

The conclusions reached by the analyst in the report are:

"When poured at an initial grout temperature of 40° C, the maximum grout temperature criterion of 90° C is not exceeded. In addition, the base radiolytic heat generation rate of 260 curies/m³ can be increased by 35% ..."

The initial pour temperature must be maintained in a certain range to meet multiple criteria. If the temperature range can be kept large, better control can be given for the peak cure temperature. Uncertainties in the rate of hydration heat generation result from variations in the waste materials (e.g. aluminum) fed into the process, and these uncertainties lead to variations in the peak cure temperature. The activity of radioactive materials have minor effect upon the peak curing temperature and are considered negligible during this stage; while the thermal conductivity and thickness of the grout and vault materials affect the rate of heat loss and thus the peak temperature.

The feed materials specification should be stated in terms of mass of heat generating materials. However, since the only materials composition and evaluation method known to be acceptable is that used in the Allen (1990) analysis, no other mix can be safely allowed unless it will generate less heat

of hydration and/or thermal analysis demonstrates waste specific acceptability.

7.2 Isotopic Mix (Radiolytic Heat Generation)

The isotopic mix fed into the grout process (Hendrickson, 1990) must also be controlled to assure that the maximum temperature will not be exceeded. The technique of Hendrickson (Hendrickson, 1991a) is an excellent way to normalize all significant contributors to a single value that can be used as a process control limit.

Hendrickson simplified the analysis of isotopic heat by excluding all isotopes that are expected to be present in low concentrations or contribute an insignificant amount of heat. However, future grout feed mixtures may include a different inventory than that determined (Hendrickson, 1990) from analysis of three tanks. The contribution of individual isotopes to the radiolytic heat generation should be included in the analysis that verifies conformance to the specified limit, unless it can be shown that the contribution is insignificant (< 0.1%). The analysis of Allen (1990) suggests, as stated by the author, that the radiolytic heat limit might be increased 35%. But the satisfactory effects of this change must be verified before it can be accepted.

The addition of 35% more radiolytic heat may require simultaneous addition of more material which in turn may affect the heat of hydration, and thus create a mix that exceeds the peak temperature criteria. The correct heat of hydration, for the actual feed associated with 35% greater radiolytic heat, must be determined and evaluated using analysis such as that of Allen (1990) with the TAPA code.

7.3 Volume Expansion

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The volume expansion (Hendrickson, 1990) may vary if the feed materials vary. The only volume expansion assessed was 1.43x for the specific conditions in the Allen (1990) analysis. Any value below 1.43 will result in higher concentrations of radionuclides and, thus, higher radiolytic heat loads. Lower mix ratios of dry materials to waste would decrease operational (hydration) heat loads.

A different feed specification would be necessary for any volume expansion factor other than 1.43. Lower values will require reduction in the radiolytic heat generating materials; a higher value would allow an increase in the radionuclide content but may be restricted by operational temperature acceptance.

7.4 Grout Thermal Conductivity

The thermal conductivity of the grouted waste may be the most critical parameter. Higher values will allow the grout to cool faster; lower values will increase the peak temperature. The minimum value 0.45 Btu/hr ft°F is much lower than that used in the Allen (1990) analysis (0.53 Btu/hr ft°F). Higher peak temperatures would be calculated if the minimum value were used in the analysis. Thus other changes in the composition of the feed would be

necessary to compensate for this lower thermal conductivity. If in fact the minimum value is the true thermal conductivity to be expected, an analysis comparable to Allen (1990) must be completed and the feed composition may require adjustment to achieve an acceptable waste feed specification and peak temperature.

7.5 Grout Vault Design

The grout vault design (Allen, 1990) also has an important effect on the peak temperature. The analytical model must be representative of the actual vault design to assure accurate temperature predictions. Conversely, changes in the design could allow greater heat loss rates and thus lower peak temperatures.

7.6 Alpha Sources

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Since alpha-emitting nuclides have a high ¹³⁷CsmBa heat equivalent (7.4 Heat Equivalents Ci/Ci), their concentration in the waste must be kept low. All alpha emitters were neglected in the evaluated analyses (Allen 1990 and Hendrickson 1990), as they are expected to be present only in very low concentrations. The expectation of low alpha emitting nuclide concentrations is derived from waste analyses (Hendrickson 1990) and by requiring that such concentrations fall below NRC class C disposal limits [(NRC 1982), 10 CFR §61.55].

At 100 nCi of total alpha per gram of waste, the waste is below the TRU limit (some alpha due to uranium), and the total contribution to the heat generation would be less than 0.5% of that found of an equivalent concentration of 260 Ci of 137 Cs per 137

The waste to be grouted must be below a TRU limit of 100 nCi/g.

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8.0 WASTE ACCEPTANCE CRITERIA AND BASES

The concentrations of some tank wastes may fall outside of the expected range defined in the source term determination. Exclusion of known incompatible waste constituents or chemicals that may prevent the grout from meeting regulatory limitations can be controlled through pre-characterization efforts and blending operations. This section defines the range of chemical compositions that are deemed to be acceptable feed to the grout facility.

8.1 Limitations Imposed by Compositional Variability

The success of the Grout Project (a stabilization/solidification process) depends on feed physical conditions and chemical characteristics. In general, a grout formulation for a specific feed is considered acceptable to meet solidified grout properties if appropriate tests indicate successful performance. The following section discusses the affects of feed physical conditions and chemical characteristics on achieving successful grouting.

8.1.1 Physical Affects

The physical conditions of the feed affect the solidification process significantly. Particle size and shape, solids content, specific gravity, temperature and other physical factors have definite affects on curing/setting and solidified grout properties. Some of the major affects from physical properties of the feed are discussed below.

Particle Size and Shape

Particle characteristics affect the viscosity of the waste and determine its rheology. Therefore, pumping/handling of the waste may be affected by the particle characteristics. Particle characteristics of the waste may also affect aspects of the solidification reactions and product homogeneity after curing (Conner 1990).

<u>Solids Content</u>

The total solids in the grout waste feed will affect the physical properties of the solidified grout and the setting/curing process because of particle sedimentation. In general, high solids content will lead to better grout curing/setting and final monolith physical properties.

Specific Gravity

Phase separation can result from large differences in the specific gravities of the feed and the reagents.

8.1.2 Chemical Composition Affects

The chemical composition of the feed to be grouted has a major impact on the setting/curing rate, physical properties of the solidified grout, and whether the mixture will even solidify. Chemicals and combinations of chemicals in the waste feed can retard, inhibit, accelerate curing/setting, and can negatively or positively affect the final grout properties of compressive stress, permeability, leachability, and others.

The effects of chemicals and combinations of chemicals in all proportions on grouting (and other solidification/stabilization processes) cannot be

predicted without appropriate verification testing for wastes not characterized by the data in Section 5.

Specific chemical factors affecting grouting of untested wastes were listed by Conner (1990) and are included in Tables 8-1 and 8-2. Chemicals that are potential problems have been identified (NRC/NISTIR 1989). The discussion that follows includes chemicals that can cause problems (experienced at other facilities grouting radioactive wastes) in grouting, potential impacts, and actions required prior to grouting.

Chemical constituents that require identification and evaluation for potential pretreatment prior to cement solidification.

- Ammonia
- Organic Acids
- Nitrates
- Phosphates
- Borates
- Chelates
- Sulfates

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- Aromatic Oils
- Soaps/Detergents

Chemicals that at ppm concentrations are known to cause problems to cement solidification operations and product acceptance and must be minimized or precluded from waste streams unless specific counteractive steps are taken.

- Acetone
- Benzene
- Hexane
- Nitrates
- Toluene

Chemicals that are known to cause problems to solidification operations and product acceptance unless characterized/quantified and appropriate formulations are used.

- Potassium Permanganate (KMnO_x)
- Paint Thinners
- 0ils
- Boric Acid Loaded on Ion Exchange Resins

There are families of chemicals that should be regarded as potentially incompatible with certain wastes and solidification formulations. The Grout facilities chemical control program and administrative procedures should be used to preclude or minimize their introduction (in uncharacterized quantities) into the waste feed.

- Hydrocarbons
- Solvents
- Petroleum Products/Lubricants
- Decontamination Solutions
- Detergents
- Oxidizing Agents

The following chemicals have created problems with solidification of radioactive waste in the past. The problems occurred when the concentrations were high and trace quantities are not a concern.

- Dry cleaning solvents. (e.g., TCE)
- Sodium Hypochlorite (NaClo)
- Ammonia
- · Ionic Soaps
- 0ils

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• Industrial Cleaners

Chemicals found to have created problems with heat generation and grout setting.

- Aluminum (dissolved) heat generation
- Sodium Fluoride (NaF) setting

8.2 Solidified Product Criteria

The solidified grout product should meet certain criteria as presented in Table 6-6 (NRC 1989). Table 6-6 includes test results for grout made from TK-241-AN-106, PSW, and simulated DSSF.

TABLE 9-1: Factors Affecting Solidification

Compound or Fector	Effect	Mechanism Affected	Chemical Fixation/ Solidification Processes
Fine particles	1,P	Р	PC, PZ
ion exchange materials	I,A	1	Al
Metal lattice substitution	F,A	1	Al
Gelling agents	R	1,2	P,I,M AI
Organics, general	I.P.A	۵,۱	Al
Acids, acid chlorides	ρ.	1	Al. Same O
Alconois, glycols	я,р-	I,W	Al. Same O
Aldehydes, ketones	р.	1	C. Some O
Amides	Я	i,W	Same O
Amines	R.A	I,F	Same O
Carbonyle	R	I.D	AI
Chlorinated hydrocarbons	ዶ. ጸ	1, M	PC, Same O
Ethers, epoxides	ρ.	t	Same O
Greate	I,P	P	PC, PC/PZ, L
Heterocyclics	p.	t	c
Hydrocarbons, general	۶.	1	C. Same O
Lignine	t	c	AI
Oil	I, P	P	PC. PZ
Starchee	1	c	Ai
Suifonates	R	0	Al
Sugare	1,8	c -	Al
Tennine	1	c	Al
Organics, specific			
Ethylene glycol	ρ	4	₽C

TABLE 8-1: Factors Affecting Solidification

	<u>-</u>	Mechanism	Chemical Fixation/ Solidification
Compound or Factor	Effect	Affected	Processes
p-Bramophenol	P-, P +		PC
Hexachioropenzene	P-,P+	1	PC
	p.	1	PC/PZ
7	P+,P-		t.
Phenoi	p. p.	1	PC, PC/PZ, L'
Frichioroethylene	P-	ı	PC, PC/PZ, L
Inorganics, general			
Acids	p.	1	PC, Same O
Bases	P. .	1	PC/SS, C, Some O
Borates	R	М	PC, PŽ
Calcium compounds			
Chlandes	R,P	1	Al
Chromium compounds	A	1	Al
Heavy metal saits	PA.R	t	Al, Some O
iron compounds	A	F,M	PC.PZ
Lead compounds	Я	M	PC.PZ
Magnesium compounds	R	M	PC.PZ
Salts, general	P-,A,R	1	Al. Some O
Silicas	R	F	PC.PZ
Sodium compounds	1	I	Al
Suifates	A.P	1 .	Al
Tin compounds	R	M	PC.PZ
norganics, specific			
Calcium chloride	A.R	M	PC,PZ
Copper nitrate	P+	1	PC
	P+,P-	1	PC/PZ
	ρ.	<u>'</u> .	L
Gypsum, hydrate Gypsum, semihydrate	A A	1	PC,PZ PC,PZ
Gypsum, seminydrate Load nitrate	Р.	1	PC.P2
road Juriefa,	P- P+	 	PC/PZ. L
Sodium hydroxide	P+.P-	ı	PC, PC/PZ, L
Sodium suifate	P-	1	PC
	P+,P+	i	PC/PZ, L
Zinc nitrate	p.	1	PC
	P+,P-	ı	PC/PZ. L

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Key effect: 🛤 setting/curing inhibition (long term); A 🗢 setting/curing acceleration; R 🛪 setting/curing retardation (short term); P+ 🛪 alteration of properties of cured product, positive effect; P- = alteration of properties of cured product, negative effect. Mechanism: P = coats particles; I = interferes with reaction; C = complexing egent; M = disrupts matrix; F = flocculent; D = dispersent; W = wetting egent, Process; PC = Portland cement-based; PC/SS = Portland cement/soluble silicate; PC/PZ = Portland cement/pozzolan; PZ = pozzolanic (kiln dust, flyesh); C = ciay-based; L = lime-based; Al = all inorganic; O = organic.

Note: When the effect may be positive or negative, depending on concentration, the first symbol listed represents lower concentration, the last higher concentration.

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TABLE 8-2: Substances Affecting Cement Reactions: Inhibition and Property Alteration

Substance or Factor	Inhibition	Property Alteration
Fine particulates	X	Х
Clay	X	
Silt	X	
Ion exchange materials	X	
Metal lattice substitution	X	
Gelling agents	X	X
Organics, general	X	X
Acids, acid chlorides	,	X
Alcohols, glycols	X	X
Aldehydes, ketones		X
Carbonyls	χ	
Carboxylates	X	
Chlorinated hydrocarbons	X	X
Grease	X	X
Heterocyclics		X
Hydrocarbons, general		X
Lignins	X	
Oil	X	Χ
Starches	X	
Sulfonates	X	
Sugars	X	
Tannins	X	
Organics, specific		
Adipic acid		
Benzene		
EDTA		
Ethylene glycol		X
Formaldehyde		
p-Bromophenol		
Hexach1orobenzene		X
Methano1		

TABLE 8-2: Substances Affecting Cement Reactions: Inhibition and

Substance or Factor	<u>Alteration</u> Inhibition	Property Alteration
NTA		
Phenols	X	· X
Trichloroethylene		X
Xylene		
Inorganics, general		
Acids		X
Bases		X
Borates	Χ .	
Calcium compounds Anions that form insoluble Ca salts		
Chlorides	X	X
Copper compounds	X	
Heavy metal salts	. Х	X
Hydroxides, insoluble		•
Hydroxides, soluble		
Lead compounds	X	
Magnesium compounds	X	
Phosphates	X	
Salts, general	X	X
Silicas	X	
Sodium compounds	X	
Sulfates	X	X
Sulfides	X	
Tin compounds	X .	
Zinc compounds	X	
norganics, specific		
Calcium chloride	X	
Copper hydroxide	X	
Copper nitrate		X
Gypsum, hydrate	X	

TABLE 8-2: Substances Affecting Cement Reactions: Inhibition and

Prop Substance or Factor	<u>perty Alteration</u> Inhibition	Property Alteration
Lead hydroxide	X	<u> </u>
Lead nitrate	X	X
Sodium arsenate	X	
Sodium borate	X	
Sodium hydroxide		X
Sodium iodate	X	
Sodium sulfate		X
Sulfur	X	
Tin		•
Zinc nitrate		X
Zinc oxide/hydroxide	X	

8.3 Limitations Imposed by Regulatory Limits

Waste feed specifications can also be identified by regulatory requirements. LDR restrictions limit the concentrations of specific wastes in the waste stream and identify constituents for which pretreatment may be necessary. Organic contaminant restrictions under LDR must be met as grouting is not currently an acceptable treatment for these constituents. The concentration of organics and toxic metals (As, Bc, Cd, Cr, Po, Hg, Se, Ag) in the waste feed are limited by the need to show compliance with TCLP testing as solidification and stabilization is the preferred treatment option for these contaminants.

For this study, EPTOX and TCLP tests of actual grout formulations were used to define probable acceptable limits. The processes of EPTOX or TCLP leaching were assumed to follow a linear trend and the recommended limits for the toxic metals were calculated from the observed test ratios (measured concentration in waste feed: measured concentration in EPTOX/TCLP extractant). This assumption is considered to be conservative because the leachate concentration of individual metal species are expected to be governed by solubility limits at a given pH rather than by initial inventory.

8.4 Heat Generation

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A thermal analysis of the vault design, the blend of materials fed to the grout process and isotopic mix must always demonstrate a peak temperature of less than or equal to 90°C, and the TRU content of the waste feed must be less than or equal to 100 nCi/gram.

8.5 Waste Feed Acceptance Criteria

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The waste feed acceptance criteria for Hanford grout are listed in Table 8-3. The final criteria are based on a comparison of limits imposed by existing regulations, heat generation (thermal limits), or compositional variability. In general, the types of organics that can be present in the waste feed is influenced by LDR guidance; the total amount of organic carbon in the waste feed (TOC) is based on documented grout production data. The amount of toxic metals in the specification is derived from EPTOX and TCLP testing. The activity of radionuclides in the waste feed is primarily determined by heat considerations. The acceptable concentrations of other cations and anions in the waste feed are based on compositional trends proven in actual tests. Specifications for some elements that are expected to be present in the waste feed (Be, Bi, Ce, La, Sb, Pd, Ta, Ti, U, V, W, Zr, CN, U, Np, Pu, Am, and Cm) could not be defined because of insufficient data. Analyses of waste constituents in product grouts will be used to define acceptance criteria for those constituents currently identified as "To Be Determined (TBD)", and to refine acceptance criteria for other constituents.

Table 8-3 provides a guide to developing grout blending strategies. The table lists the maximum concentration of waste constituents that probably will not result in a violation of existing regulations for TCLP metals, will conform to previously tested grout mixtures, and are not likely to cause heat generation concerns due to radionuclide decay. However, care must be taken to ensure that the <u>proportions</u> of constituents are consistent with previous grout tests to avoid synergistic effects that might impact grout cure-time, strength, or rheology.

The waste feed acceptance criteria are based on the most current information available. In some instances, a specified limit may not be the maximum concentration that can be successfully grouted. As the grout program develops, additional data will become available and should be incorporated in the waste feed acceptance table as appropriate.

The regulatory-based limits in the first column of Table 8-3 are based on the test results from Section 6. The values in the first column were derived assuming that the regulatory limit is proportional to the feed concentration as shown below for arsenic:

Arsenic regulatory-based limit in feed

$$= \frac{0.03 \ (Table \ 6-1 \ for \ DSSF)}{\langle 1.0 \ (Table \ 6-7, EPTOX)} * 5 \ (Table \ 6-7 \ Reg. \ Limit) = 0.15$$

<u>TABLE 8-3:</u>	Grout F	eed Acce	otance	Criteria

Feed Component	Regulatory- Based Limit	Thermal Limit	Proven Groutability	Acceptable Limit	Acceptance Criteria Reference
Organics (ppm)					
TOC	-	-	1556	1556	1,6,7
Other Organics	Toxicity limits for individual organic species must be determined on a case- by-case basis (see Table 4-2)	-	-	See Table 4-2	14
Cations/Metals	(mqq)				
Ag	5063	-	162	5063	2,6,7
AT	•	-	20300	20300	2,6,7
As	0.15	-	0.03	0.15	2,6,5
8	-	-	136	136	6,7
Ba	46154	-	600	46154	2,6,7
Ве	-	-	-	-	TBD-WM-004
Bi	-	•	-	-	TBD-WM-005
Ca	•	-	573	573	6,7
Cd	80	-	8	80	2,6,5
Ce	•	-	•	-	TBD-WM-006
Cr	21000	-	1260	21000	2,6,7
Cu	-	-	7	7	6,7
Fe	-	-	1490	1490	6,7
Hg	20	-	3	20	2,6,5
K	-	-	11500	11500	6,7
La	-	-	-	-	TBD-WM-007
Li	-	-	-	-	TBD-WM-008
Mg	- .	-	320	320	6
Mn	-	-	3010	3010	6,7
Мо	-	-	68	68	6,7
Na	-	-	122000	122000	2,6,7
Nd	. -	-	-	•	TBD-WM-009
Ni	-	-	30	30	6

Feed	Regulatory-	Thermal	Proven	Acceptable	Acceptance
Component	Based	Limit	Groutability	Limit	Criteria
	Limit	·			Reference
Pb	12.5	-	2.5	12.5	2,6,7
Pd	-	-	-	-	TBD-WM-010
Sb	•	-	-	-	TBD-WM-011
Se	45	-	4.5	45	2,5
Si	-	-	502	502	2,6
Ta	-	-	-	•	TBD-WM-012
Ti	-	-	-	-	TBD-WM-013
U	-	-	-	•	TBD-WM-014
٧	-	-	•	-	TBD-WM-015
W	-	-	-	• •	TBD-WM-016
Zn	-	-	2930	2930	6,7
Zr		-	-		TBD-WM-017
Anions (ppm)					
C1	-	-	5360	5360	3,6,7
CN (free)	-	-	-	-	TBD-WM-018
CN (total)	-	-	-	-	TBD-WM-019
CO ₃	-	-	22920	22920	6,7
F ⁻	-	-	562	562	3,6,7
NO ₃	-	-	186000	186000	3,6,7

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	TABLE 8-	3: Grout	Feed Acceptance	<u>Criteria</u>	
Feed	Regulatory-	Thermal	Proven	Acceptable	Acceptance
Component	Based	Limit	Groutability	Limit	Criteria
	Limit				Reference
Radionuclides	(Ci/L)				4,12
H-3	l6 μCi/L	•	-	16 μCi/L	14
C-14	0.647	-	-	0.647	11
Co-60	-	0.1162	-	0.1162	4,12
Se-79	80.6	-	-	80.6	11
Sr-90	10.01	0.2662	-	0.2662	4,12,13
Nb-94	120.7	-	-	120.7	11
Tc-99	0.2617	•	-	0.2617	11
Ru-106	-	0.1855	-	0.1855	4,12
Sb-125	•	0.5399	-	0.5399	4,12
I-129	0.00107	-	-	0.00107	11
Cs-134	-	0.1761	•	0.1761	4,12
Cs-137	6.578	0.3718	-	0.3718	4,12,13
Ce-144	-	0.2237	-	0.2237	4,12
U-234	•	•	-	-	
U-235	-	•	-	-	12,
U-238		-	-	-	TBD-WM-014
Np-237		-	-		
Pu-238	Total TRU		-	Total TRU	
Pu-239/240	concentra- tion <100	-	-	concentra- tion <100	11,13
Am-241	nCi/g	-	-	nCi/g	
Cm-244		•			
Other Paramete	rs				
pH (Standard Units)	-	-	-	>10	14
Total Solids (ppm)	-	-	-	<400,000	14
Heat Generators	-	<0.26 CsmBa heat equiv. Ci/L	-	<0.26 CsmBa heat equivalents Ci/L	4,12
Density	-	-	<u>.</u>	< 1.4 Kg/L	14

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Notes: (All concentrations expressed as weight percent unless noted.)

- Total organic constituents should not exceed 1556 mg/L.
- 2. Total sodium (Na) should be >75% of total cations. Total aluminum (Al) should be <20% of total cations. Waste specifications for As, Ba, Cd, Cr, Pb, Hg, Se, and Ag based on EP toxicity and TCLP tests assuming linearity between waste feed concentration and extract concentrations.
- 3. Total nitrate-nitrite (NO_3-NO_2) should be <75% of total anions. Total chloride-fluoride-hydroxide-carbonate $(Cl-F-OH-CO_3)$ should be less than 20% of total anions.
- 4. Concentrations based on Hendrickson (1991a).
- 5. Use of higher regulatory limit is not expected to compromise groutability of waste.
- Lokken et al., 1989.
- 7. Serne et al., 1989a.
- 8. Serne et al., 1989b.
- 9. DOE Order 5400.5.
- 10. Whyatt, 1991.

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- 11. Performance goal is to limit maximum individual exposure from grout through all pathways to 5 mrem/yr or 0.8 mrem/yr from drinking water (Whyatt, 1991) as a summation of dose consequences, such would include 137 Cs and 79 Se. Individual contributors calculated as the 95% confidence mean concentration divided by the Performance Assessment Table 4-2 Base Case Fraction of Performance Goal.
- 12. The total mix of radionuclides in the grout feed must be evaluated to assure that the net concentration in CsmBa equivalent curies is below 260 per m³. The evaluation method is based on the sum of the fractions rule as described in Hendrickson (1990).
- 13. NRC, 10 CFR §61.
- 14. Specific organics basis 40 CFR §268. Tritium basis air emission permits (Hendrickson 1991b). pH basis tank compatibility. Total solids and density bases equipment compatibility.

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APPENDIX A: WASTE SOURCE TERM DATA

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 $\frac{\text{TABLE }A-1}{\text{Source Term Concentrations for Organics (ppm)}}$

Organics	Mean Value¹	95% ² Confidence	Bounding ³ Source Term
TOC	2300	5672	14616
N-C ₂₂ H ₄₆ - N-C ₄₀ H ₃₂	2.8	10.9	32.4
N-C ₂₂ H ₄₆ - N-C ₃₄ H ₇₀	1.4	5.4	16.2
Alkyl, hydroxymethyl benzene	0.17	0.7	2.0
Methyltoluidine	0.33	1.3	3.8
n-Dimethyltoluidine	1.1	4.3	12.8
2-Chloromethy1, hydroxymethy1benzene	1.2	4.6	13.5
2-Chloromethyl-o-xylene	0.62	2.5	7.4
Ethylxylene	0.03	0.1	0.4
Ethyl, 2-methyl hydroxymethylbenzene	4.4	17.0	50.6
2-Methylhydroxymethyl benzene	33	129	384
C ₃ -alkylbenzene	30	118	350
Propylbenzene	0.17	0.7	2.0
Trimethylbenzene	7.3	29.2	87.4
Ethylbenzaldehyde	65	250	742
Methylbenzaldehyde	65	250	742
Diethylphthalates	0.94	3.6	10.8
Unknown phthalates	2.7	7.6	20.6
Dioctylphthalates	2.5	8.7	25.3
Chloroethyl, 2-hydroxymethyl benzoic acid	1.2	4.6	13.5
2-Hydroxymethylbenzoic acid	2.5	10.0	29.7
2-Methylbenzoic acid	1.7	6.6	19.6
Butanedioic acid	39	154	458
n-Dodecane	0.61	1.6	4.1
Dodecanoic acid	0.13	0.5	1.5
EDTA	340	1301	3850
ED3A	3	9.9	28.2

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TABLE A-1
Source Term Concentrations for Organics (ppm)

Organics	Mean Value ¹	95% ² Confidence	Bounding ³ Source Term	
HEDTA	1300	5177	15463	
MICEDA	2.9	11.2	33.1	
MAIDA	54	211	627	
Ethanedioic acid	390	1536	4577	
Hydroxyacetic acid	800	3160	9421	
NTA [nitriloacetic acid]	1.5	4.2	11.4	
Heptadecanoic acid	0.23	0.9	2.6	
Heptanedioic acid	2.6	10.0	29.7	
Hexadecanoic acid	0.12	0.5	1.4	
Hexanedioic acid	7	23.2	66.1	
Hexanoic acid	4.1	15.9	47.2	
Octadecanoic acid	0.058	0.2	0.7	
n-Pentadecane	0.46	1.4	3.7	
Pentadecanoic acid	3.3	12.9	· 38.4	
Pentanedioic acid	6.6	25.1	74.3	
Tri-n-butyl phosphate	5.5	15.3	41.2	
[(Tri-n-butyl)di-ol] phosphate	1.1	4.1	12.2	
Citric acid	1400	5615	16795	
n-Tetradecane	1.9	4.8	12.4	
n-Tridecane	3.4	8.6	22.5	
n-Undecane	0.52	1.8	5.3	

Notes: Ref: Hendrickson (1990)

- 1. Mean Value: The mean concentration of DSSF waste from tanks 241-AW-101, 241-AN-103, and 241-AN-106.
- 2. 95% Confidence (95% Confidence Interval Limit): The concentration that represents the upper limit of the one-tailed 95% confidence interval for the data distribution exhibited by samples from tanks 241-AW-101, 241-AN-103, and 241-AN-106.

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3. Bounding Source Term: The source-term concentrations used for design analyses, safety analyses etc. Bounding source term is based on mean concentration; sample standard deviation; and probability factors. The probability factors describe observed data distribution and tolerance limits that quantify the likelihood that source-term concentrations measured in subsequent sampling events will not exceed those previously observed at a particular confidence interval.

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TABLE A-2
Source Term Concentrations for Cations/Metals (ppm)

Cations/Metals	Mean Value ¹	95% ² Confidence	Bounding ³ Source Term
Ag	4.3	8	17.8
AI	12000	18406	35400
As	29	67.8	171
В	4.7	18.4	54.6
Ba	4.6	7.1	13.8
Be	5.5	7.7	13.5
Bi	76	186	476
Ca	36	64.7	141
Cd	12	30.5	79.7
Ce	12	47.4	141
Cr	300	620	1470
Cu	3.5	7.0	16.4
Fe	15	25.5	53.2
Нд	2.3	7.7	22.0
K+	7000	21498	59959
La	0.1	0.4	1.2
Li	1.9	7.5	22.2
Mg	7.1	16.7	42.2
Mn	7.2	16.5	41.1
Мо	26	27.9	32.8
Na+	100000	112138	144338
Nd	4.3	16.9	50.5
Ni	21	54.7	144
Pb	63	176	. 476
Pd	9.3	36.3	108
Sb	55	76.9	135
Se .	22	75.9	219
Si	49	87.8	191

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TABLE A-2
Source Term Concentrations for Cations/Metals (ppm)

Cations/Metals	Mean Value ¹	95% ² Confidence	Bounding ³ Source Term
Ta	43	169	505
Ti	4.5	7.7	16.2.
U	29	54.3	121.4
٧	5.5	7.7	13.5
W	61	67.6	85.0
Zn	9	15.6	33.0
Zr	33	111	316

TABLE A-3
Source Term Concentrations for Anions (ppm)

Anions	Mean Value¹	95% ² Confidence	Bounding ³ Source Term
C1 -	2700	3105	4178
CN (free)	0.0038	0.0148	0.044
CN (total)	21	. 51.3	132
CO ₃	7900	24084	67017
F-	290	813	2199
NO ₃ -	78000	145435	324320
NO ₂ -	34000	43272	67869
OH-	27000	53974	125528
PO ₄ =	4200	15495	45459
SO ₂ =	1500	3186	7658

Source Term Concentrations for Radionuclides (Ci/

Source Term	<u>Concentrations</u>	for Radionucli	des (Ci/L)
Radionuclides (Ci/L)	Mean Value ¹	95% ² Confidence	Bounding ³ Source Term
H-3	7.0 E-06	1.6 E-05	3.90 E-05
C-14	8.4 E-07	1.1 E-06	1.83 E-06
Co-60	1.1 E-05	2.8 E-05	7.20 E-05
Se-79	6.7 E-06	2.5 E-05	7.44 E-05
Sr-90	6.6 E-03	1.1 E-02	2.32 E-02
Nb-94	1.0 E-05	3.5 E-05	1.02 E-04
Tc-99	7.7 E-05	8.9 E-05	1.22 E-04
Ru-106	4.3 E-03	1.7 E-02	4.99 E-02
I-129	1.7 E-07	3.0 E-07	6.56 E-07
Cs-134	1.2 E-03	4.7 E-03	1.41 E-02
Cs-137	3.1 E-01	3.7 E-01	5.26 E-01
U-234	1.2 E-08	3.2 E-08	8.59 E-08
U-235	7.0 E-10	2.1 E-09	5.75 E-09
U-238	8.2 E-09	1.6 E-08	3.65 E-08
Np-237	5.8 E-08	2.1 E-07	6.00 E-07
Pu-238	4.3 E-07	.8.0 E-07	1.78 E-06
Pu-239/240	9.0 E-07	1.7 E-06	3.92 E-06
Am-241	1.4 E-06	2.0 E-06	3.56 E-06
Cm-244	7.7 E-08	2.4 E-07	6.87 E-07

TABLE A-5

Other Parameters	
рН	>10
Total Solids	300 g/L (2.5 lb/gal)
Specific Gravity	1.3

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